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This grant was used to purchase a Coherent 400-15 Innova laser and a Mira 900 Dual Cavity Mode Locked Ti-Sapphire laser for use in our research on investigating the optical properties of photorefractive crystals.

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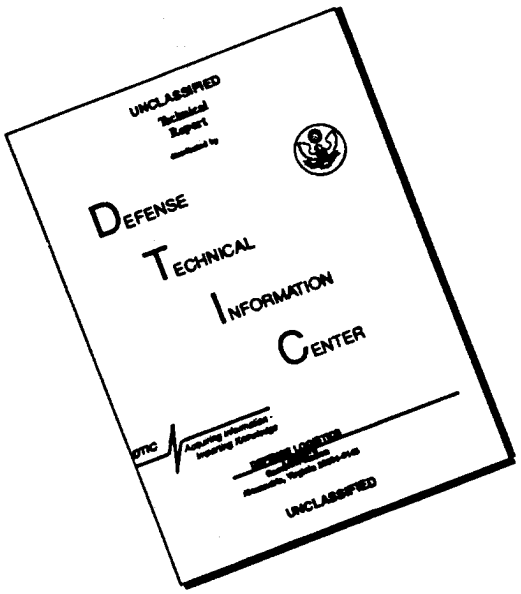

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Final Report

Grant F49620-95-1-0079 (Account 61031)

"Instrumentation for Recording Optically Induced Dynamic Ferroelectric Domain Gratings & Generating Quasi-Phase Matched, Tunable Second Harmonic Light"

Principal Investigator: Amnon Yariv
California Institute of Technology

This grant was used to purchase a Coherent 400-15 Innova laser and a Mira 900 Dual Cavity Mode Locked Ti-Sapphire laser for use in our research on investigating the optical properties of photorefractive crystals.

We have used the allotted $113,200, obtained a trade-in of $45,241 for our old lasers and shared in the cost of the equipment with Caltech funds of $12,269.

The equipment is now being used routinely to investigate the optical properties of photorefractive crystals with special emphasis on inducing ferroelectric periodic domain structures (with emphasis on applications to data storage and second harmonic generation).

Financial Summary:

1.	Coherent, Innova 400-15, Argon Ion Laser	$65,366.00
2.	Coherent, Mira 900, Dual-Cavity Mode-Locked Titanium Sapphire Laser	$95,000.00
	Less trade-in of obsolete CIT owned lasers (2)	$45,241.00
	Sales Tax	9,497.81
	Shipping	846.58
	TOTAL	$125,469.39
Amount Charged To Grant		$113,200.00
Amount Paid With Institute Funds		$12,269.39

(Cost sharing consisted of $12,269.39 in Institute plus $45,241.00 in Institute-owned equipment which was traded in.)

Optically induced quasi-phase matching in strontium barium niobate

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We observe dynamic ferroelectric domain gratings in strontium barium niobate (SBN) induced by photorefractive space charge fields. The optically induced modulation of the spontaneous polarization attains a maximum of 1%. Quasi-phase matched second harmonic enhancements are observed above the ferroelectric-paraelectric phase transition due to the glassy ferroelectric nature of SBN. We find that the second harmonic power is significantly enhanced by recording gratings in optically fatigued rather than electrically poled crystals. © *1995 American Institute of Physics.*

We have recently demonstrated that ferroelectric domains align with the local photorefractive space charge fields in $Sr_xBa_{1-x}Nb_2O_6$(SBN:x).[1,2] The resulting dynamic domain gratings respond nearly instantaneously to changes in the photogenerated space charge field and modulate the ferroelectric polarization. We used these gratings to perform tunable quasi-phase matched second harmonic generation (QPM-SHG) with spectral widths of the QPM enhancement as narrow as 0.175 nm, across the fundamental tuning range of 880–990 nm. In this letter we apply this technique to SBN:61 and explore the temperature dependence of the phenomenon. We demonstrate that space charge field induced QPM-SHG can be achieved even in the paraelectric phase. In addition, we observe that an optically fatigued crystal displays significantly stronger QPM-SHG than an electrically poled crystal. We also find that a strong broad-band second harmonic enhancement occurs automatically in optically depoled crystals.[3,4] In fact, this broad-band enhancement can attain values several orders of magnitude larger than the QPM enhancement. We propose an explanation of this latter result based on charge compensation and fringe stability requirements.

The experimental setup for writing dynamic domain gratings and simultaneously generating the second harmonic is identical to that reported in Ref. 2. A tunable, mode-locked Ti-sapphire laser is frequency doubled within the 45° cut, Ce-doped SBN:61 crystal (4.5×4.5×5.5 mm). This crystal exhibits a ferroelectric-paraelectric phase transition at 75 °C. The fundamental infrared beam is focused to a 60 μm beam diameter, producing a peak fundamental intensity of 17 MW cm^{-2} at the beam waist. A 2–6.6 W argon-ion laser at 514.5 nm records a domain grating throughout the entire crystal volume. To maintain the poled state at elevated temperatures, a poling field of 8000 V cm^{-1} is applied between optical exposures (Fig. 1).

The angular spectrum of the second harmonic wave reveals the spatial modulation of the spontaneous polarization and nonlinear optical susceptibility.[5–7] Upon recording a domain grating in an initially poled crystal, we observe a collimated QPM second harmonic beam [Fig. 1(a)], in addition to a non-phase matched streak. This streak exits the crystal at an angular displacement of approximately 10° below the QPM spot. The origin of this angular walk-off between the two distinct second harmonic beams is the 45° inclination of the c axis relative to horizontal [Fig. 1(b)]. The broad extent of the streak along the x direction and the narrow extent in the y direction indicates that domains are extremely narrow in diameter, while being elongated along the c axis. Typical dimensions of microdomains are tens of nanometers in diameter and hundreds of nanometers in length. Figure 1(b) depicts the resulting range of grating vectors generated by the randomly distributed microdomains, represented as a multiplicity of grating vectors with different magnitudes directed nearly normal to the c axis and all sharing a common origin. The momentum conservation relation for the interaction is $2\mathbf{k}^{\omega}+\mathbf{k}_g=\mathbf{k}^{2\omega}$, where $\mathbf{k}^{\omega}$ is the fundamental wave vector and $\mathbf{k}^{2\omega}$ is the second harmonic wave vector. The second harmonic far field profile will be directed to a spot (upon interacting with grating periodicity $\mathbf{k}_g$) and a streak (upon interacting with the ensemble of grating periodicities $\mathbf{k}_{\text{random}}$). For a random distribution of microdomains of diameter 10 nm, the distribution of $\mathbf{k}_{\text{random}}$ is also random up to grating vectors as large as 6×10^8 m^{-1}.

The total second harmonic power in the streak can attain

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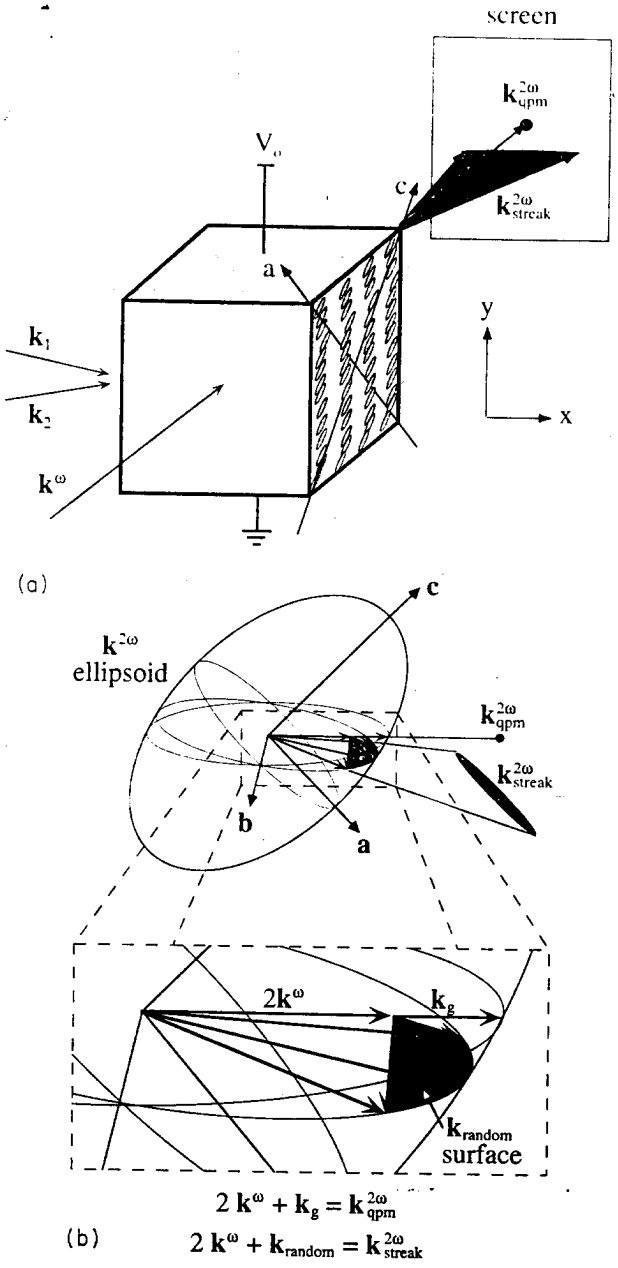

FIG. 1. (a) Experimental setup: Optical waves $\mathbf{k}_1$ and $\mathbf{k}_2$ record the dynamic domain grating. $\mathbf{k}^{\omega}$ is the fundamental beam and the second harmonic waves exit the crystal to form both a spot $\mathbf{k}^{2\omega}_{qpm}$ and a streak $\mathbf{k}^{2\omega}_{streak}$ in the far field. (b) k space diagram illustrating the origin of the relative angular displacement of second harmonic spot (QPM contribution, $\mathbf{k}_g$) and streak (random domain contribution, $\mathbf{k}_{random}$). The random domain grating vectors contributing to the streak are indicated by the shaded $\mathbf{k}_{random}$ surface.

a value two orders of magnitude stronger than the QPM spot if the random depolarization of the crystal is more severe than the periodic domain inversion. The random depolarization arises because each time a QPM grating is recorded, inverted microdomains form along those grating planes in which the space charge field is directed opposite to the spontaneous polarization. Instabilities in the spatial phase of the optical interference pattern (arising from optical heating, for instance) tend to uniformly depole the crystal over time. Thus, the volume of uniformly distributed, inverted microdomains continues to increase as multiple holograms are exposed. In contrast, the domain modulation at the periodicity of the hologram compensates the instantaneous photorefractive space charge field. This spontaneous polarization modulation depth is small in comparison to the broadband distribution of periodicities arising from the accumulation of microdomains. As a result, the broadband second harmonic conversion efficiency is enhanced by a factor of 1000 (to 1%) in an optically depoled crystal, above the conversion in a poled crystal. Long term fringe stability (greater than hours) is necessary to maximize the periodic domain modulation while minimizing the uniform depolarization. In practice, this level of stability is extremely difficult to achieve.

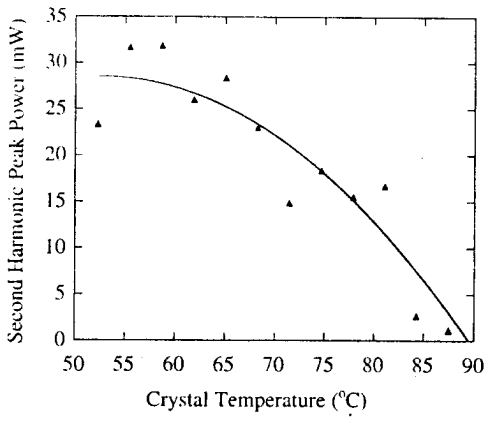

FIG. 2. Temperature dependence of quasiphase matching peak enhancement for gratings written with total intensity of 1 W cm^{-2} at 514.5 nm. Note the existence of QPM-SHG above the Curie temperature $T_c = 75$ °C.

We next focus on the periodic depolarization generated by the photorefractive space charge field. By heating the crystal to within 20 °C of the ferroelectric phase transition, we find that the QPM enhancement, or equivalently the modulation of the spontaneous polarization, attains a maximum. Furthermore, the glassy ferroelectric property of SBN is manifest in the observation of QPM-SHG above the ferroelectric phase transition temperature (Fig. 2). We believe the origin of this effect is the alignment of micropolar regions in the paraelectric phase under the influence of the space charge field. In fact, micropolar regions are believed to exist up to temperatures as high as 300 °C.[8] However, the QPM enhancement observed here becomes extremely weak at temperatures in excess of 85 °C.

The second harmonic power is dramatically enhanced by recording gratings in optically fatigued crystals; that is, crystals which have been exposed to significant optical energy ($>10^4$ J cm^{-2}) at 514.5 nm [Fig. 3(a)]. The formation of random microdomains in the illuminated regions reduces the macroscopic spontaneous polarization by a factor of 3 to 10 (as indicated by the degradation of the electrooptic coefficient). The preexistence of microdomains facilitates the subsequent domain grating formation. The small volume of an individual microdomain (<100 nm^3) lowers the activation energy required to invert the dipole.

The second harmonic power arising from a single coherence length ($l_c = 1.12$ μm) slice of the crystal is predicted to be 0.1 mW from published values of the nonlinear optical susceptibilities.[9] In our measurements, the background sec-

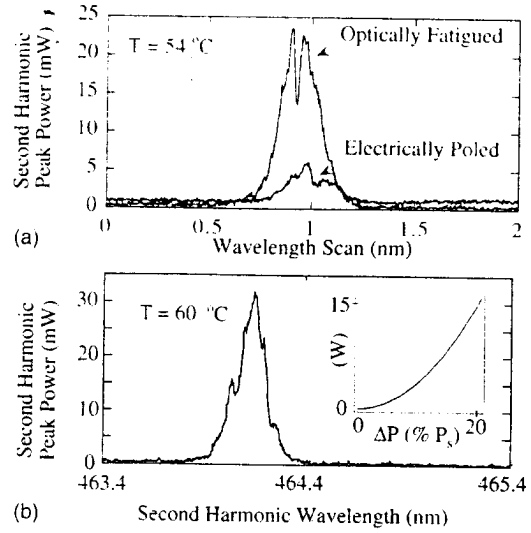

FIG. 3. (a) Enhancement of second harmonic power in an optically fatigued crystal following a high intensity exposure for ~3 h, 1 W cm^{-2}. The noise features superimposed on peak are due to fluctuations in the interference pattern. (b) Typical QPM tuning curve in SBN:60 at 60 °C for 3.25 μm grating written with a total intensity of 1 W cm^{-2}. Maximum conversion efficiency is 0.01%. Inset: calculated second harmonic peak power as a function of the percent modulation of the spontaneous polarization (for a 17 MW cm^{-2} fundamental plane wave).

ond harmonic is ~1 mW, indicating that the conversion efficiency is enhanced by the broad-band random domain contribution. This power is centered on the QPM spot and does not include the power contribution from the second harmonic streak. From the second harmonic power at the QPM peak [Fig. 3(b)] we estimate the depth of periodic modulation of the spontaneous polarization. For instance, the measured peak second harmonic power of 30 mW corresponds to an effective crystal length of 40 coherence lengths. The spontaneous polarization is equivalently modulated by 1%, so an effective nonlinear optic susceptibility induced by the space charge field is approximately 0.06 pm V^{-1}. To attain conversion efficiencies of several percent at these fundamental beam intensities (17 MW cm^{-2}), the spontaneous polarization must be modulated by at least 20%. The inset of Fig. 3(b) illustrates the predicted second harmonic power as a function of the periodic polarization modulation.

We believe that the modulation of the spontaneous polarization during relatively short optical exposures (typically less than 15 min) is limited by the mobile charge available to compensate the depolarization fields at head-to-head domain walls. The carrier density required to compensate the nonzero divergence of the polarization, for a grating composed of domain walls whose surface normal is at 45° to the c axis, is given by:[10]

$$N_{\text{comp}}=\sqrt{2}\,\frac{\pi\Delta P}{q\Lambda_g}, \tag{1}$$

where ΔP is the polarization modulation amplitude at spatial periodicity Λ_g and q is the charge of an electron. The carrier density required to produce a field equal to the coercive field is

$$N_{\text{coercive}}=\frac{2\sqrt{2}\,\pi\epsilon E_{\text{coercive}}}{q\Lambda_g}, \tag{2}$$

where q is the charge of an electron and ϵ is the low frequency dielectric constant. For P_s equal to 25 μC cm^{-2}, E_{coercive} equal to 1 kV cm^{-1} (typical for SBN:61 at 45 °C) and a grating period equal to 3.25 μm, the compensating charge density required on the domain walls for *bipolar* modulation ($\Delta P=2P_s$) is $N_{\text{comp}}\sim 10^{18}$ cm^{-3}, and the coercive charge density is 4×10^{16} cm^{-3}. The charge should be localized about the domain walls, so in fact significantly higher charge densities may be required (perhaps a factor of 10 larger). In the experiments described here, the small effective modulation of the spontaneous polarization (1%) indicates that the spatially periodic photoinduced space charge density is 10^{16} cm^{-3}–10^{17} cm^{-3}, depending on the degree of localization of the compensating charge. This suggests that further improvement in the QPM second harmonic power can be attained by tailoring the effective density of photorefractive traps to increase the space charge field while not dramatically changing the absorption at the second harmonic wavelength.

In conclusion, we have found that domain gratings can be recorded above the ferroelectric-paraelectric phase transition due to the glassy ferroelectric nature of SBN, and the QPM-SHG is significantly enhanced by recording gratings in optically fatigued rather than electrically poled crystals. The QPM-SHG is significantly weaker than the broadband second harmonic enhancement arising from the microdomain structure. We believe that the relatively small QPM-SHG conversion efficiencies are a consequence of the limited compensating charge available to screen the depolarization fields. Therefore, the primary effort to increase the domain modulation should be directed at increasing the effective density of photorefractive traps through optimization of the dopant chemistry.

The authors would like to acknowledge the support of ARPA, the Air Force Office of Scientific Research, and ONR.

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